

Unusually high critical current of clean P-doped BaFe₂As₂ single crystalline thin film

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Microstructurally clean, isovalently P-doped BaFe₂As₂ (Ba-122) single crystalline thin films have been prepared on MgO (001) substrates by molecular beam epitaxy. These films show a superconducting transition temperature (T_c) of over 30 K although P content is around 0.22, which is lower than the optimal one for single crystals (i.e. 0.33). The enhanced T_c at this doping level is attributed to the in-plane tensile strain. The strained film shows high transport self-field critical current densities (J_c) of over 6 MA/cm² at 4.2 K, which are among the highest for Fe based superconductors (FeSCs). In-field J_c exceeds 0.1 MA/cm² at $\mu_0 H = 35$ T for $H \parallel ab$ and $\mu_0 H = 18$ T for $H \parallel c$ at 4.2 K, respectively, in spite of moderate upper critical fields compared to other FeSCs with similar T_c . Structural investigations reveal no defects or misoriented grains pointing to strong pinning centers. We relate this unexpected high J_c to a strong enhancement of the vortex core energy at optimal T_c , driven by in-plane strain and doping. These unusually high J_c make P-doped Ba-122 very favorable for high-field magnet applications.

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Electron and hole doped BaFe₂As₂ (Ba-122) compounds show high upper critical fields with low anisotropy at low temperatures.¹ These properties are favorable for high-field applications and, indeed, high-performance K-doped Ba-122 wires have been fabricated by a powder-in-tube technique.^{2,3} Although, the growth of partially oriented K-doped Ba-122 thin films prepared by a two-step process has been presented⁴, in-situ K-doped Ba-122 thin films are difficult to realize due to the high vapor pressure of K. It has been reported that K-doped Ba-122 thin films were prepared by molecular beam epitaxy (MBE), although the films are not stable in air.⁵ On the other hand, isovalently P-doped Ba-122 thin films with a superconducting transition temperature (T_c) of around 30 K, which is the second highest T_c in the Ba-122 family, have been fabricated by both pulsed laser deposition (PLD) and MBE, where films prepared by the former method showed slightly lower T_c .⁶⁻⁸ Additionally, bicrystal experiments revealed a high critical current density (J_c) of around 1 MA/cm² even at a grain boundary angle as large as 24° at 4 K,⁶ far beyond the superconducting properties of Co-doped Ba-122.⁹⁻¹¹

Hence, P-doped Ba-122 may be a good candidate for Ba-122 wires and tapes applications. However, only properties at relatively low fields (up to 9 T) have been investigated so far. In order to use this material class for

applications, the knowledge of in-field transport properties in a wide range of external fields and orientations needs to be extended. Here, we present high-field (dc up to 35 T) transport properties of a P-doped Ba-122 epitaxial thin film prepared by MBE and discuss the feasibility of applications for this material class.

The P-doped Ba-122 thin film has a thickness of 107 nm, obtained from transmission electron microscopy (TEM) and was grown on a MgO (001) substrate by MBE. The substrate was clamped to the holder with a thin indium layer in between to achieve high thermal conductivity from the heater. All elements were supplied from Knudsen cells using solid sources: Ba, Fe, As, and GaP (GaP decomposes and produces a Ga-free film). The detailed fabrication process of the thin film can be found in Ref. 12. Electron probe micro analysis was used to determine the film stoichiometry and it results to be Ba:Fe:(As+P) = 1 : 1.92 : 1.93 with a P content of P/(P+As) = 0.22, which is lower than the optimal doping level for single crystals (i.e., 0.33).^{13,14} X-ray diffraction shows the c -axis oriented growth of the film with high phase purity (Fig. 1a). The (103) Φ -scan (Fig. 1b) and the pole figure measurements (not shown in this letter) of the film revealed an in-plane orientation with a full width at half maximum (FWHM) value $\Delta\phi$ of 0.56°. Rocking curve measurements on the (004) reflection show a sharp peak with a FWHM value $\Delta\omega$ of 0.54° as can be seen in Fig. 1c. (Values are not corrected for device broadening) The epitaxial relation was confirmed to be (001)[100]Ba-122||[(001)[100]MgO. The overview bright-field TEM image (Fig. 1d) was taken on an FEI Tecnai

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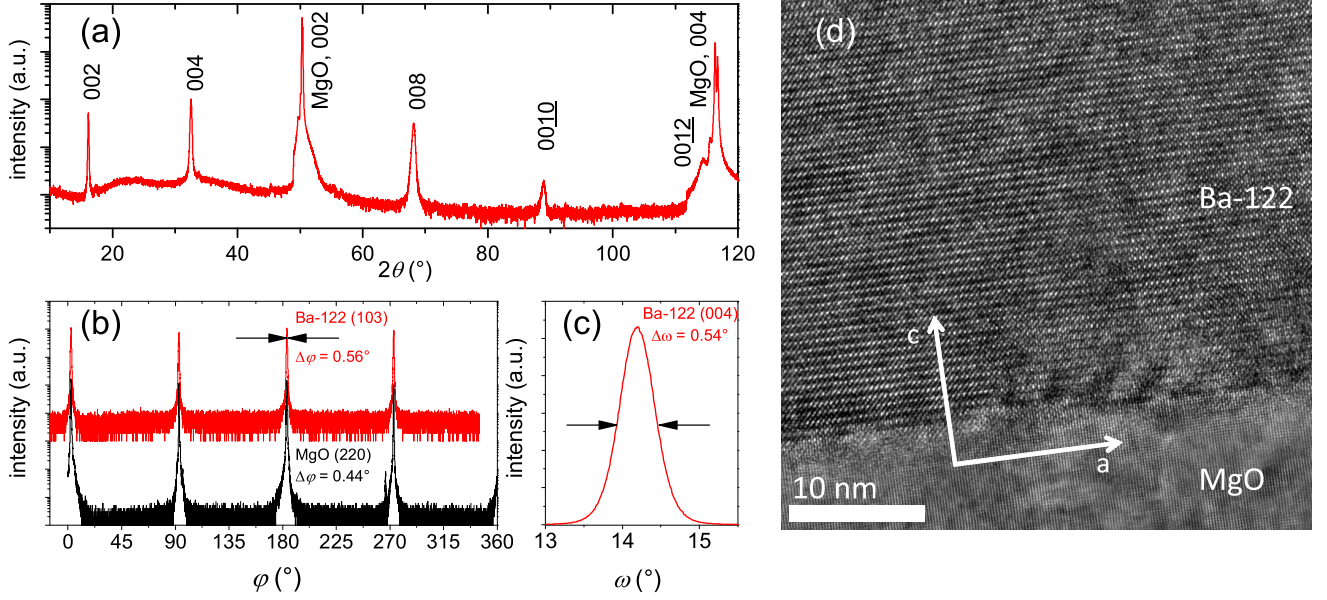


Figure 1: Structural data of the P-doped Ba-122 thin film. (a) $\theta - 2\theta$ scan showing the c -axis oriented growth. (b) ϕ -scan of the (103) reflection of the Ba-122 and the (220) reflection of the MgO substrate proving the cube-on-cube film growth. (c) Rocking curve of the (004) reflection (d) TEM image near the interface between the MgO substrate and the Ba-122 phase.

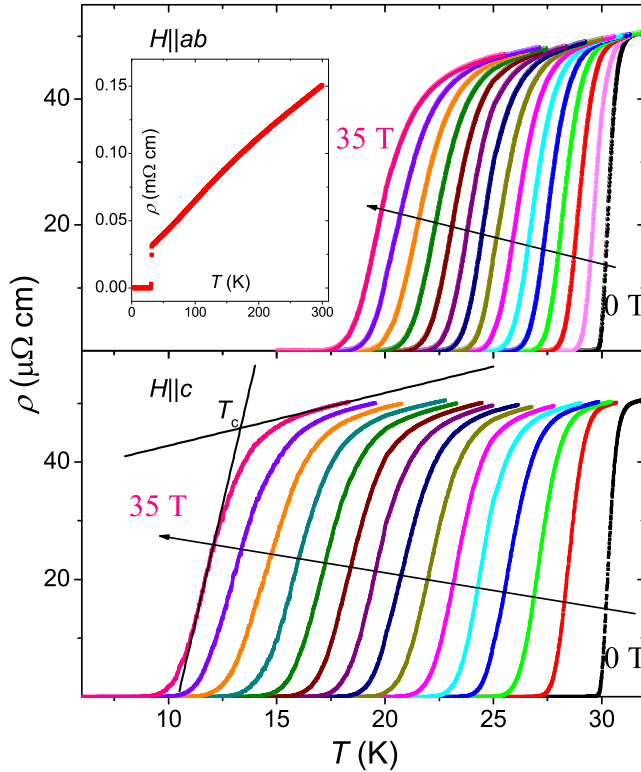


Figure 2: Field dependence of the resistive transition for the P-doped Ba-122 thin film for $H \parallel ab$ (top) and $H \parallel c$ direction (bottom). The field steps are 2.5 T. The inset shows the temperature dependence of the resistivity from 2 K up to 300 K in zero field.

T20 (LaB₆, 200 kV). High-resolution imaging was carried out using an FEI Titan 80-300, operating at 300 kV (field emission gun) with an image C_s corrector. The lamella was prepared with the in-situ lift-out method in a focused ion beam device.¹⁵

In Fig. 1d no reaction layer can be seen at the interface between P-doped Ba-122 thin film and MgO substrate. Additionally, neither appreciable defects nor grain boundaries were observed in TEM investigations (lower magnifications images are not shown in this letter), indicating high crystalline quality and phase purity of the investigated films.

High field transport measurements on a small bridge of 1 mm length and 40 μ m width, prepared by laser cutting, were conducted in the dc facility up to 35 T at the National High Magnetic Field Laboratory (NHMFL) in Tallahassee, FL, at 4.2 K. A criterion of 1 μ V/cm was used to define J_c . The higher temperature measurements were performed in a 16 T Physical Properties Measurement System. The magnetic field H was applied in maximum Lorentz force configuration during all measurements ($H \perp J$, J being the current density). Resistivity measurements in the absence of magnetic field unveiled an onset T_c of 30.7 K, which is higher than that of PLD processed films^{8,16} and comparable with the T_c of optimally doped single crystals.¹⁷ However, the film has a lower P content than optimally doped single crystals. The lattice parameter $c = 12.78$ Å of our film (evaluated from XRD analysis presented in Fig. 1a, using the Nelson-Riley fit) is shorter than that of single crystals with the same P content ($c = 12.88$ Å).¹³ This arises from the presence of tensile strain in our films. It has been reported that

tensile strain in the underdoped regime for P-doped Ba-122 enhances T_c .¹² The normal state resistivity shows a linear temperature dependence below 100 K (inset of Fig. 2), which is a typical behavior for optimally P-doped Ba-122.¹⁸ Therefore, the tensile strain shifts the superconducting transition of our film to higher values¹² comparable to a T_c of higher P concentrations without strain, a dependency which has been observed for Co-doped Ba-122 thin films as well.¹⁹

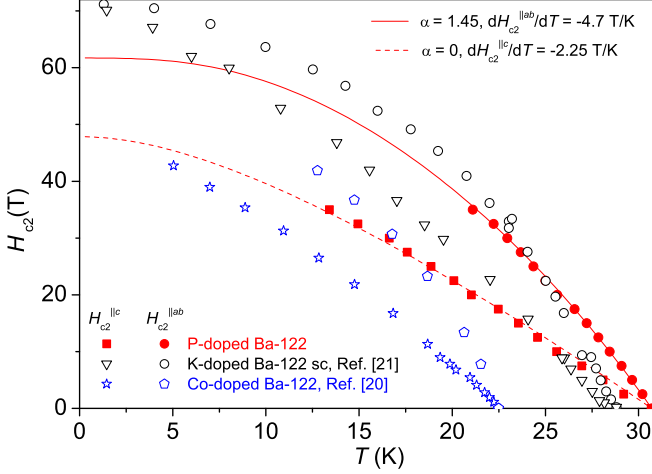


Figure 3: Temperature dependence of the upper critical field of P-doped Ba-122 (red filled symbols) in $H||c$ (squares) and $H||ab$ (circles) direction. The data for a Co-doped Ba-122 thin film²⁰ (blue stars and pentagon) and a K-doped Ba-122 single crystal²¹ (black triangles and circles) of comparable T_c are shown as well. The red lines are the single-band WHH fits of the P-doped Ba-122.

The temperature dependence of the resistivity $\rho(T)$ was measured in magnetic fields up to 35 T in both main crystallographic directions (Fig. 2). The small broadening of the transition width with increasing magnetic field is similar to other Ba-122 systems, due to the small Ginzburg number.²² The temperature dependence of the upper critical field (H_{c2}) values evaluated from the resistivity data shown in Fig. 2 is plotted in Fig. 3. The H_{c2} data for both directions (i.e., $H||c$ and $H||ab$) can be fitted by the single-band Werthamer-Helfand-Hohenberg (WHH) model.²³ The temperature dependence of H_{c2} for $H||c$ does not show any paramagnetic limiting effects up to 35 T in accord with literature data¹⁷ (i.e., Maki parameter $\alpha = 0$). However, H_{c2} for $H||ab$ is reduced at low temperatures compared to an extrapolation from the slope at T_c with $\alpha=0$. In this case we obtained a value of $\alpha = 1.45$. This can be attributed to Pauli-limiting behavior which is typical for FeSCs.^{24,25} As can be seen in Fig. 3, $H_{c2}(T)$ for P-doped Ba-122 is relatively small compared to other compounds having comparable T_c .^{20,21,26} Additionally, the H_{c2} anisotropy of P-doped Ba-122 is larger than that of Co-doped and K-doped Ba-122. In general, the critical current densities are very high for P-

doped Ba-122 compared to other FeSCs, as shown below.

In Fig. 4a the temperature dependence of the self-field J_c for our P-doped Ba-122 film is compared to other J_c results reported for FeSCs. It is seen that P-doped Ba-122 shows the highest self-field J_c values among other FeSCs. Note, that even SmFeAs(O,F) (Sm-1111) with a much higher T_c of 54 K shows smaller J_c values.^{27–29} Our clean P-doped Ba-122 film presents a self-field J_c of about 6.3 MA/cm² at 4.2 K, which is almost 7% of the depairing current density.^{22,30} We assume that this J_c could be further increased by adding additional pinning centers.⁷ As can be seen in Fig. 4a, the film prepared by the same method but with an excess of Fe content during the growth has twice the value of J_c .⁶ The field dependence of J_c at various temperatures is summarized in Fig. 4b. The J_c values for $H||c$ ($J_{c,H||c}$) are always lower than those for $H||ab$, and no feature close to $H||c$ is observed in Fig. 4c: these findings indicate the absence of c -axis correlated pinning and that the material anisotropy dominates the general J_c trend. The pinning force density (F_p , insets of Fig. 4b) calculated according to $F_p = H \times J_c$ at 4.2 K shows values up to 77 GN/m³ (at 15 T) for $H||ab$. The data for $H||c$ show a maximum of around 35 GN/m³ at around 10 T. Compared to the results presented by Miura et al.^{7,8} and Adachi et al.¹⁶, our film showed slightly higher values which might be due to the higher T_c . In particular J_c at 35 T $H||c$ is as high as $J_{c,H||c} = 1 \cdot 10^4$ A/cm². In general, J_c of optimally P-doped films is quite robust against applied magnetic fields. The question arises why our microstructurally clean film exhibits such high J_c values. Usually, a high density of defects is necessary to achieve high J_c . However, we did observe neither crystal structure defects nor impurity phases in our films (Fig. 1). Alternatively, it has been shown by Putzke et al. that the vortex core energy of the flux lines is enhanced close to the optimal doping.¹⁷ Therefore, we suppose that this high vortex core energy is a key factor responsible for the unusually high J_c in optimally P-doped Ba-122. In this context, comparable J_c values of the P-doped Ba-122 with T_c around 25 K containing BaZrO₃ particles⁷ or strong pinning centers³¹ could be explained by the reduction of the vortex core energy due to non optimal T_c .

Figure 4c shows the angular dependence of J_c [$J_c(\theta)$] measured at 4.2 K and various fields up to 35 T. J_c has a broad maximum positioned at $\theta = 0^\circ$ ($H||ab$) and no prominent J_c peaks at $\theta = 90^\circ$ ($H||c$). Low $J_c(H)$ -anisotropy values ($\gamma_{J_c} = J_{c,H||ab}/J_{c,H||c}$) of around 2 up to 15 T at 4.2 K are observed, increasing for higher fields. Noteworthy is the observation of a small shoulder near the ab -peak, marked with arrows. It shifts to lower angles when the magnetic field is increased. Such shoulders are known from cuprates and exist usually due to strong correlated defects, such as seen in double-perovskite-doped YBa₂Cu₃O₇ (YBCO) thin films³², or due to uncorrelated extended defects, such as in Au-irradiated YBCO thin films³³ or in Sm_{1+x}Ba_{2-x}Cu₃O_{7-d} thin films with Sm-rich precipitates.³⁴ There, the shoulders are accompanied

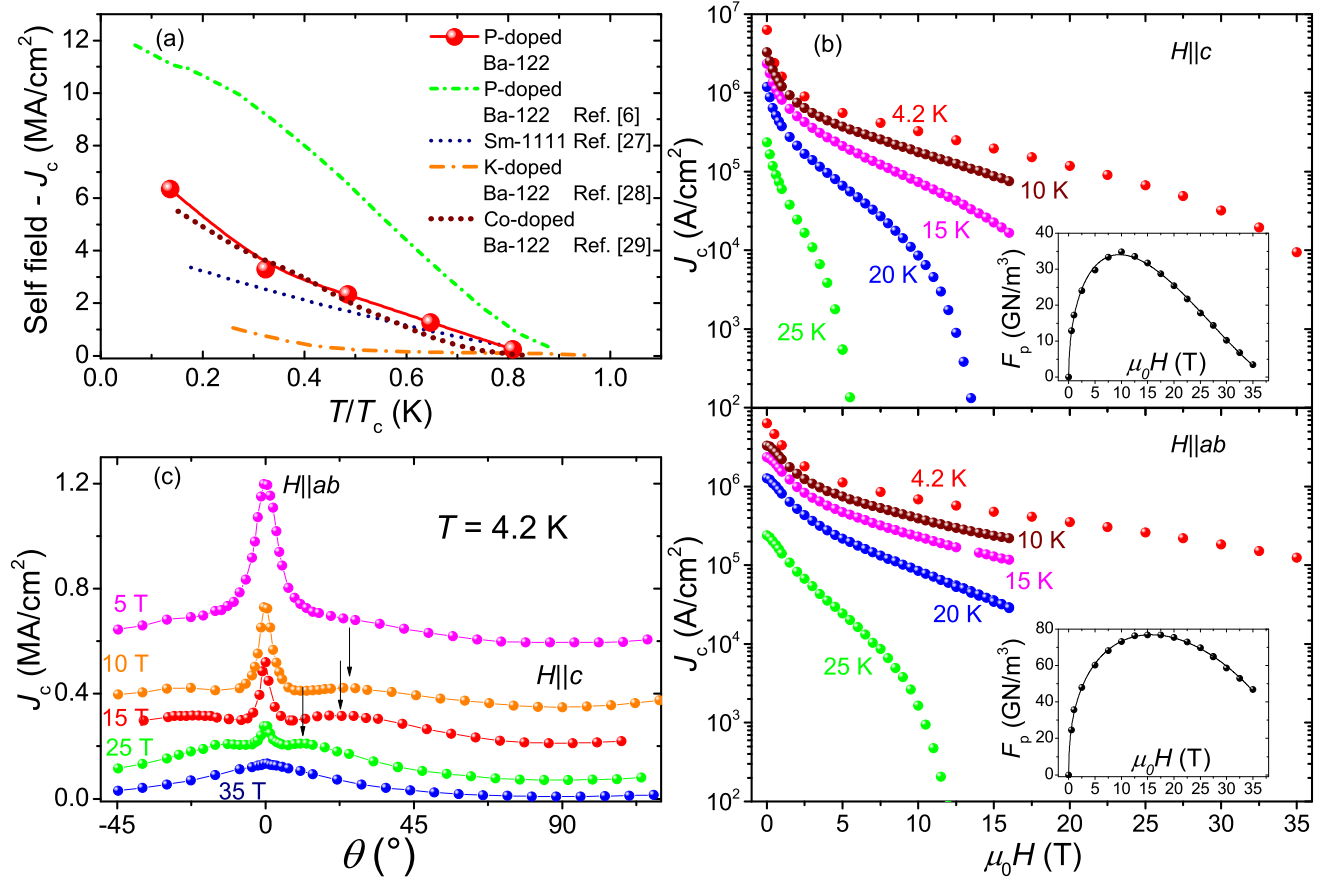


Figure 4: (a) Self-field J_c in dependence on the normalized temperature for several FeSCs.^{6,27–29} The temperature is normalized to $T_{c,onset}$ for Sm-1111 to $T_{c,90}$. (b) Field dependence of J_c of P-doped Ba-122 for both major crystallographic directions. The insets show the respective pinning force densities at 4.2 K. (c) Angle dependence of J_c for different applied fields at 4.2 K.

by extended structural defects and often also by a strong c -axis peak in a certain magnetic field range. For our films, we did not observe any extended or correlated defects in TEM images nor any sign of off-axis peak by XRD. Therefore, the reason for these shoulders may lie in a possible variation of the P-content. For example, nanoscale regions of non-optimal P-content (not observable in TEM) of size slightly larger than the coherence lengths might act as uncorrelated strong pinning centers due to the unusually strong P-content dependence of the vortex core energy.^{17,35} These possible P-inhomogeneities in combination with the inequality of λ and ξ anisotropy as in the FeSCs can lead to such shoulders, as was shown by van der Beek et al.³⁶ The presence of the pinning centers is further evidenced by the $J_c(T)$ dependence at intermediate and high temperatures (Fig. 4a). Additional artificial disorder in the form of Fe impurities enhances the strong pinning resulting in a $J_c(T)$ dependence consistent with the δI pinning scenario in the whole temperature range.³⁷ Thus, we believe pinning driven by differences in the vortex core energy enhances J_c values of the optimally P-doped Ba-122 well above all other FeSCs.

There is still room for further increase in J_c of the P-doped Ba-122 thin films, like doping with pinning-promoting particles the way it was done by Miura et al.⁷ in combination with optimal growth^{31,38} and high crystalline quality. Additionally, a high concentration of a secondary phase like Fe can be incorporated into the superconducting matrix as artificial pinning centers for J_c increase without detrimental decrease in T_c .⁶ This implies that the J_c anisotropy can be reduced while maintaining high T_c as well as J_c . The unusually high critical currents makes the P-doped Ba-122 one of the most promising materials among FeSCs for the study of the superconducting pairing mechanisms and high field applications.

To conclude, using MBE we have fabricated P-doped Ba-122 thin film directly on MgO achieving epitaxy and phase purity with a high T_c of 30.7 K. We measured the field and the angle dependence of J_c up to 35 T. A very high self field J_c of 6.3 MA/cm² at 4.2 K was observed even though no structural defects were found in TEM and XRD. This observation suggests that in the optimally doped P-doped Ba-122 compound rather weak structural

inhomogeneities result in strong pinning centers. This unusual pinning enhancement is explained by a sharp maximum in the vortex core energy near to the optimal doping.

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